

Spin Dependent Collision of Ultracold Metastable Atoms

Satoshi Uetake,^{1,2,*} Ryo Murakami,¹ John M. Doyle,³ and Yoshiro Takahashi^{1,2}

¹*Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*

²*CREST, Japan Science and Technology Agency, Chiyoda, Tokyo 102-0075, Japan*

³*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*

(Dated: March 8, 2013)

Spin-polarized metastable atoms of ultracold ytterbium are trapped at high density and their inelastic collisional properties are measured. We reveal that in collisions of $\text{Yb}(^3P_2)$ with $\text{Yb}(^1S_0)$ there is relatively weak inelastic loss, but with a significant spin-dependence consistent with Zeeman sublevel changes as being the dominant decay process. This is in strong contrast to our observations of $\text{Yb}(^3P_2)\text{--Yb}(^3P_2)$ collisional loss, which are, at low field, much more rapid and have essentially no spin dependence. Our results give a guideline to use the 3P_2 states in many possible applications.

Atoms with alkaline-earth-metal-like electronic structure are under extensive study, partly due to their promise for a number of key applications. For example, the use of long-lived metastable P states (as well as the ground S state) has been explored as a useful quantum computing platform [1–5]. Also, the ultranarrow $^1S_0\text{--}^3P_0$ atomic resonance in a “magic wavelength” optical lattice may be highly competitive as a new optical frequency standard [6]. Finally, in the area of quantum simulation, there are several theoretical studies of the use of 3P_J ($J = 0, 2$) atoms for studies of Hamiltonians with both spin and orbital degrees of freedom [7, 8], implementation of Abelian artificial gauge potentials [9], or simulation of Kondo lattice model [10].

Collisions of metastable P state atoms are interesting not only from a fundamental interactions viewpoint, and as the determining factor in collisional cooling schemes, but also as a crucial mechanism in several key applications. Quantum gate phase imprinting via collisions has been proposed [2, 5, 11], as well as the exchange interaction in Kondo lattice model simulations [10]. Collisions can not only enable new physics but also inhibit desired applications. For example, in optical frequency standards collisional shifts can limit their accuracy, as well as lead to inelastic losses that destroy the atomic sample, possibly on a time scale shorter than the desired interrogation time. Recent studies with metastable alkaline-earth-metal-like structure atoms showed inelastic collision rates as high as 10^{-17} to 10^{-16} m^3/s in $^3P_J\text{--}^3P_J$ collisions in Yb [12] and Sr [13, 14] atoms. These rates, almost as high as the estimated elastic collisional rates, make the use of 3P_J atoms difficult.

The role of the collisions between the ground (1S_0) and excited (3P_J) atoms is crucially important in many approaches [2, 5, 10]. In this respect, it is important to investigate the properties of collisions between the metastable triplet states and the ground state [14, 15]. In addition, the study of anisotropically interacting cold collisions is now a broadening area of study and includes the rare-earth atoms and polar molecules [16–24].

In this Letter, we report the measurement of the inelastic rate constants for Yb in both the $^3P_2\text{--}^3P_2$ and

$^1S_0\text{--}^3P_2$ systems at several magnetic fields below 1 gauss. In particular, we observe strong spin dependence in the inelastic rates for $\text{Yb}(^3P_2)$ in the $^1S_0\text{--}^3P_2$ collisional system, with higher energy Zeeman sublevels having higher inelastic rates. This strongly suggests that the inelastic loss is dominated by Zeeman sublevel changing processes (“ m -changing collisions”). These may be induced by the Landau-Zener transition between the entrance s -wave channel and the higher partial waves with lower magnetic sublevels [25, 26]. In contrast, in the $^3P_2\text{--}^3P_2$ collisions we observe a much higher inelastic rate that is spin-independent. This is consistent with fine structure changing processes (“ J -changing collisions”) or principal quantum number changing (“PQNC”) processes. Our results represent a new, detailed study of 3P_2 physics, and also provide a roadmap for the use of the important 3P_2 state and challenge theory to provide a quantitative explanation of this anisotropic collision physics.

In a previous experiment [12], we studied 3P_2 collisions via indirect excitation of the 3D_2 state to 3P_2 . This led to a spin-unpolarized sample at the relatively high temperature of around 40 μK . Here we prepare spin polarized samples at a much lower temperature, below 1 μK , as described below. The experimental procedure is as follows. Yb atoms in a thermal beam generated from an oven at 375°C are decelerated by a Zeeman slower with a strong $^1S_0\text{--}^1P_1$ transition at 399 nm, and then are loaded into the intercombination ($^1S_0\text{--}^3P_1$) MOT at 556 nm [27, 28]. Figure 1(b)–(d) show corresponding energy level diagram of Yb, timing diagram of the experiment, and schematic diagram of experimental setup, respectively. The atoms are transferred from the MOT to a crossed FORT at 532 nm and evaporatively cooled. The number of atoms is typically 3×10^5 . The BEC transition temperature (T_c) of our trap is 400 nK. All the work reported in this paper is done at 480 nK, just above T_c , in order to maintain a stable atom number. Our spin-selective excitation employs the very narrow linewidth $^1S_0\text{--}^3P_2$ transition at 507 nm [29, 30]. The excitation efficiency to 3P_2 is about 10%, typically leaving about 2×10^4 3P_2 atoms in the trap. During the excitation we apply a small bias magnetic field B_{bias} to spectroscopically split magnetic

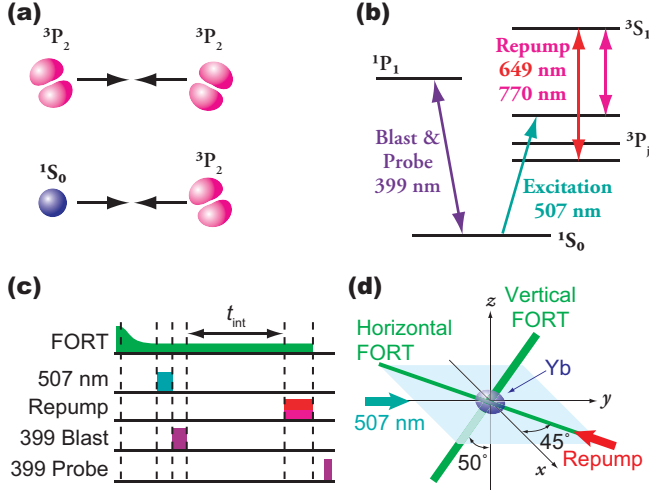


FIG. 1. (color online) (a) Schematics of the collision measurement. We studied both 3P_2 - 3P_2 and 1S_0 - 3P_2 collisional properties. (b) Energy level diagram of ytterbium. (c) Timing diagram for measurement of collision dynamics of 3P_2 - 3P_2 and 1S_0 - 3P_2 collision system. For measurement of 3P_2 - 3P_2 collision system, unexcited 1S_0 atoms are immediately removed from the trap by applying a strong 399 nm laser. For measurement of 1S_0 - 3P_2 collision, the blast laser is irradiated after interaction time. (d) Schematic diagram of experimental setup. Directions of 399 blast and probe lasers (not shown) are same as that of 507 nm excitation laser.

sublevels m_J of 3P_2 state. The applied magnetic field varies from 215 mG to 848 mG, corresponding to the Zeeman splitting of 0.45 MHz to 1.8 MHz. The excitation laser linewidth is measured to be less than 1 kHz and Doppler broadening is about 20 kHz. This is much less than the Zeeman splitting of 450 kHz at our lowest magnetic field of 215 mG, thus ensuring selective creation of spin-polarized samples of atoms in a single Zeeman sublevel.

We perform study of two different collisional systems, 3P_2 - 3P_2 and 1S_0 - 3P_2 . For the former, the leftover 1S_0 atoms are immediately removed by strong excitation at 399 nm (i.e. blast laser) [Fig. 1(c)]. After interaction time t_{int} (where the 3P_2 atoms collide with other atoms and undergoes inelastic loss), 3P_2 atoms are repumped back to the 1S_0 state by 770 nm and 649 nm repumping lasers which are resonant to the 3P_2 - 3S_1 and 3P_0 - 3S_1 transitions. A few milliseconds is required for complete repumping. Finally the number of repumped (1S_0) atoms are measured by absorption imaging using the 1S_0 - 1P_1 transition. For the latter (i.e. 1S_0 - 3P_2 collision measurement), we simply leave the leftover 1S_0 atoms in the trap. Then the blast laser is irradiated after interaction time: just before the repumping.

Inelastic atom-atom collisions in various channels are clearly observed for all Zeeman states. A selection of these state-dependent decay curves (those at highest and lowest magnetic field) are presented in Fig. 2. Figure

2a (b) shows the time evolution of the number of 3P_2 atoms without (with) the 1S_0 atoms. Thus the decay of atoms in Fig. 2a is due only to 3P_2 - 3P_2 collisions. The measurements reveal a very high inelastic loss rate (approximately the same as the estimated elastic rate) that is essentially independent of both m_J and magnetic field strength. This behavior is consistent with the scenario that the dominant decay process is J -changing, which was inferred by our previous measurement [12]. It may also be due to PQNC collisions, as described in Ref. [13].

The decay of 3P_2 state atoms in collisions dominantly with 1S_0 atoms is shown in Fig. 2b. It is important to note that in these decay curves the number of 1S_0 atoms is ten times the number of 3P_2 atoms in either Fig. 2a or 2b. Thus, although the decay curves are steeper in this raw data, as will be explained in detail later and shown in Fig. 3, 1S_0 - 3P_2 collisions are less inelastic than 3P_2 - 3P_2 collisions. One can easily recognize in Fig. 2b that the 1S_0 - 3P_2 collisional decay behavior is markedly different from that of 3P_2 - 3P_2 collisions. The observed decay curves are strongly state-dependent. In particular, the atoms in higher Zeeman levels in the 3P_2 state ($m_J \geq 0$) show a stronger field dependence in the decay. This clearly suggests the important role of m -changing collisions in the 1S_0 - 3P_2 collision.

We extract 2-body inelastic loss rate coefficients from the observed decay rates using the following method. The decay is modeled by coupled differential equations:

$$\begin{aligned} \dot{n}_e &= -\beta_{ge}n_gn_e - \beta_{ee}n_e^2, \\ \dot{n}_g &= -\beta_{ge}n_gn_e, \end{aligned} \quad (1)$$

where n denotes the local density of atoms, subscript g (e) denotes ground (excited) state, β_{ge} (β_{ee}) is the two body loss coefficient for ground-excited (excited-excited) state collisions. The trap lifetime for the ground state atoms is measured to be 30 s, thus we neglect background collision loss because it is much longer than the present two-body collision loss. For the 3P_2 - 3P_2 collision, Eqs. (1) become simple and are solved analytically, since all atoms in the ground state are removed from the trap ($n_g = 0$). By spatially integrating the equation, we obtain the evolution in atom number

$$N_e(t) = \frac{1}{1/N_{0e} + G_{ee}t}. \quad (2)$$

Here N_{0e} is the initial excited state atom number and $G_{ee} = (V_{2e}/V_{1e}^2)\beta_{ee}$. The effective volume is defined by

$$V_{qe} = \int d^3r [n_e(\mathbf{r})/n_{0e}]^q, \quad (3)$$

where q is an integer number, n_{0e} is the peak density of excited state atoms, and $n_e(\mathbf{r})$ is the spatial density distribution.

The spatial density distribution depends on the dimensionless parameter η : the ratio of the trap depth ϵ_t to

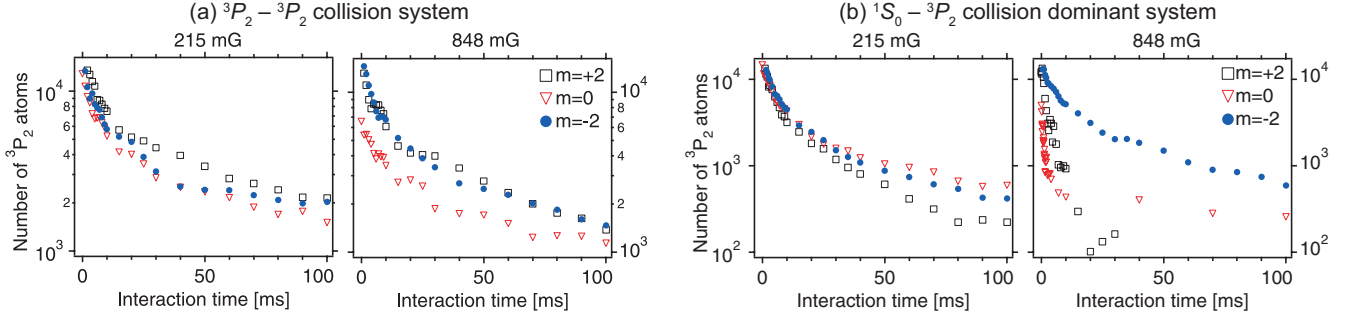


FIG. 2. (color online) Observed decay of trapped 3P_2 atoms as a function of time. (a) Time evolution of the number of 3P_2 atoms without 1S_0 atoms. Decay rates of different magnetic sublevels m_J at different magnetic field strengths are essentially the same. (b) Time evolution of the number of 3P_2 atoms with 1S_0 atoms. In this case the number of 1S_0 atoms are 10 times larger than that of 3P_2 atoms. Thus the 3P_2 atoms dominantly collides with 1S_0 atoms. The behavior is markedly different from that of 3P_2 - 3P_2 collision system. Decay rates are strongly spin-dependent. Note that we measured decay for all magnetic components in each bias magnetic field of 215, 307, 407, 596, and 848 mG. Part of data is shown in these graphs.

the sample temperature $k_B T$ in energy unit. In our experiment, η is 10 for the ground state atoms. The trap depth for each m_J component in 3P_2 state strongly depends on the direction of external magnetic field and the polarization of FORT laser [30, 31]. To determine η of the 3P_2 state, we measure the Stark shift for all m_J states as a function of horizontal FORT laser power in various bias magnetic fields studied here. The ratio U_e/U_g , where $U_{e(g)}$ is the trap potential of the 3P_2 (1S_0) state, varies from 1.02 to 1.31. From the measurement, η for all m_J states are calculated to be 7.2 to 11.7. Since η keeps more than 7 for any m_J components, large- η approximation can be used to calculate the effective volume. In the large- η limit, $n(\mathbf{r})$ is well approximated by the thermal density distribution [32], $n_e(\mathbf{r}) = n_{0e} \exp[-U(\mathbf{r})/k_B T]$, where $U(\mathbf{r})$ is the trap potential. By approximating FORT potential to a truncated harmonic trap $U(r) = \epsilon_t (r/R_0)^2 \Theta(R_0 - r)$, the effective volume can be written as

$$V_q = R_0^3 \left(\frac{\pi}{q\eta} \right)^{3/2}, \quad (4)$$

where $\Theta(x)$ is the Heaviside step function, $U(R_0) = \epsilon_t$ is the trap depth, and R_0 is the boundary.

We can extract the β_{ge} coefficient by analyzing the decay of 3P_2 atoms in the presence of 1S_0 atoms, shown in Fig. 2 (b). The data involves the 1S_0 - 3P_2 collision as well as the 3P_2 - 3P_2 . Thus, the analysis is not so simple as Eq. (2), because Eqs. (1) cannot in general be solved analytically. However, if we assume depletion of the number of ground state atoms is negligibly small, Eqs. (1) can be solved analytically. In this case, we obtain the atom number evolution in a large- η approximation

$$N_e^A(t) = \frac{\exp(-G_{ge} N_{0g} t)}{\frac{1}{N_{0e}} + \frac{1}{N_{0g}} \frac{G_{ee}}{G_{ge}} [(1 - \exp(-G_{ge} N_{0g} t))]}, \quad (5)$$

where $G_{ge} = \beta_{ge} / (V_{1e}^{2/3} + V_{1g}^{2/3})^{3/2}$ and N_{0g} is the ini-

tial ground state atom number. Note that we can reproduce Eq. (2) from Eq. (5) if we set $N_{0g} = 0$.

We first fit the 3P_2 - 3P_2 collision data by using Eq. (2) and calculate β_{ee}^{in} . Then decay data of 1S_0 - 3P_2 is fitted by using Eq. (5) with fixed β_{ee}^{in} . As mentioned in Ref. [33], the observed two-body decay rate β includes inelastic collision loss β^{in} and evaporation β^{el} due to elastic collision; i.e., $\beta = \beta^{\text{in}} + f\beta^{\text{el}}$, where f represents the fraction of elastic collisions. The inelastic collision rate can be expressed as $\beta^{\text{in}} = \beta/(f\gamma + 1)$, where γ is the ratio of elastic to inelastic cross sections, $\gamma \equiv \sigma^{\text{el}}/\sigma^{\text{in}}$ [33]. In a large- η approximation, f can be approximated to a simple analytic equation [34]. In the present condition, β^{in}/β varies from 0.87 to 0.93.

The inelastic collision rates for all m_J states in various external magnetic fields are plotted in Fig. 3. Essentially no spin-dependence in the collision of two metastable 3P_2 states is apparent, which is represented by triangles in Fig. 3. In particular, the spin states of $m_J > -2$ show almost the same inelastic collision rates with the lowest energy state of $m_J = -2$, which should not suffer from the m -changing collision. This behavior is quite different from the significant spin dependence theoretically predicted in the collision of two metastable 3P_2 state at a high magnetic field [25, 26]. Therefore, our result clearly shows that the m -changing collision is not observed for all spin states at this low magnetic field and low temperature. Since the most likely decay process for the $m_J = -2$ state is the J -changing collision from the 3P_2 state to 3P_0 or 3P_1 , this would be dominant. PQNC collision may also contribute to the overall decay. However it is difficult to distinguish J -changing from PQNC collision unless we measure β_{ee} for 3P_0 - 3P_0 collision, a task beyond the scope of this Letter.

Note that the average of the obtained inelastic collision rate coefficients β_{ee} of $4 \times 10^{-17} \text{ m}^3/\text{s}$ is a little higher than the value of $1.0 \times 10^{-17} \text{ m}^3/\text{s}$, which is previously obtained at a 100 times higher temperature [12]. This inter-

esting temperature dependence is open to a further theoretical investigation. In contrast, the significant spin- and field-dependence in the collision of the metastable 3P_2 state with the ground state 1S_0 is observed, which is represented by squares in Fig. 3. Especially, the spin states with higher energies ($m_J > -2$) show higher inelastic collision rates compared with the lowest energy state of $m_J = -2$. Since the $m_J = -2$ state differs from other spin states only in that the $m_J = -2$ state does not suffer from the m -changing collision, it is natural to think that the dominant decay process is a m -changing collision in this $^3P_2-^1S_0$ collision at a low field. The quantitative theoretical explanation of the detailed behaviors is an interesting future work [35].

In addition, we can claim that the $m_J = -2$ state is rather stable against the collision with the 1S_0 atom. Since the decay of the 3P_2 ($m_J = -2$) atoms is dominated by the $^3P_2-^3P_2$ collision even in the presence of the 1S_0 atoms, as is shown in Fig. 3, it is quite difficult to accurately extract the β_{ge} coefficient for $m_J = -2$ state. Although our analysis results in the value of β_{ge} on the order of 10^{-19} m³/s for the $m_J = -2$ state, it may be much lower. In fact, our recent measurement on $^1S_0-^3P_2$ ($m_J = -2$) atoms in a tightly confined three dimensional optical lattice indicates that the upper limit of β_{ge} is on the order of 10^{-20} m³/s [36]. This is promising to use in many applications.

In conclusion, we have experimentally investigated collisional properties of spin polarized metastable 3P_2 states of Yb Atoms. We reveal the significant spin-dependence in the collision of the metastable 3P_2 state with the ground state 1S_0 , which strongly suggests that the dominant decay process is a m -changing collision. On the contrary, we observe essentially no spin-dependence in the collision of two metastable 3P_2 states, which is consistent with the J -changing collision as a dominant decay process. Our results will trigger theoretical efforts to clarify these behaviors quantitatively and give a guideline to use the 3P_2 states in many possible applications.

The authors acknowledge very helpful experimental assistance of S. Sugawa, S. Kato, and Yb experiment team in Kyoto university. We also thank T. Tscherbul, P. Zhang, and A. Dalgarno for helpful discussions. This work was supported by the Grant-in-Aid for Scientific Research of JSPS (No. 18204035, 21102005C01 (Quantum Cybernetics), 22684022), GCOE Program “The Next Generation of Physics, Spun from Universality and Emergence” from MEXT of Japan, and FIRST. One of us, J. M. D., thanks the Fulbright program for support.

* Present address: Department of Physics, Okayama University, 3-1-1, Tsushima-naka, Kita-ku, Okayama 700-8530, Japan; uetake@scphys.kyoto-u.ac.jp

- [1] K. Shibata, S. Kato, A. Yamaguchi, S. Uetake, and Y. Takahashi, Appl. Phys. B **97**, 753 (2009).
- [2] A. J. Daley, M. M. Boyd, J. Ye, and P. Zoller, Phys. Rev. Lett. **101**, 170504 (2008).
- [3] R. Stock, N. S. Babcock, M. G. Raizen, and B. C. Sanders, Phys. Rev. A **78**, 022301 (2008).
- [4] A. Derevianko and C. C. Cannon, Phys. Rev. A **70**, 062319 (2004).
- [5] A. V. Gorshkov, A. M. Rey, A. J. Daley, M. M. Boyd, J. Ye, P. Zoller, and M. D. Lukin, Phys. Rev. Lett. **102**, 110503 (2009).
- [6] H. Katori, M. Takamoto, V. G. Pal’chikov, and V. D. Ovsinnikov, Phys. Rev. Lett. **91**, 173005 (2003).
- [7] A. V. Gorshkov, M. Hermele, V. Gurarie, C. Xu, P. S. Julienne, J. Ye, P. Zoller, E. Demler, M. D. Lukin, and A. M. Rey, Nature Phys. **6**, 289 (2010).
- [8] C. Xu, Phys. Rev. B **81**, 144431 (2010).
- [9] F. Gerbier and J. Dalibard, New Journal of Physics **12**, 033007 (2010).
- [10] M. Foss-Feig, M. Hermele, and A. M. Rey, Phys. Rev. A **81**, 051603 (2010).
- [11] D. Jaksch, H.-J. Briegel, J. I. Cirac, C. W. Gardiner, and P. Zoller, Phys. Rev. Lett. **82**, 1975 (1999).
- [12] A. Yamaguchi, S. Uetake, D. Hashimoto, J. M. Doyle, and Y. Takahashi, Phys. Rev. Lett. **101**, 233002 (2008).
- [13] A. Traverso, R. Chakraborty, Y. N. Martinez de Escobar, P. G. Mickelson, S. B. Nagel, M. Yan, and T. C. Killian, Phys. Rev. A **79**, 060702 (2009).
- [14] C. Lisdar, J. S. R. V. Winfred, T. Middelmann, F. Riehle, and U. Sterr, Phys. Rev. Lett. **103**, 090801 (2009).
- [15] N. D. Lemke, J. von Stecher, J. A. Sherman, A. M. Rey, C. W. Oates, and A. D. Ludlow, Phys. Rev. Lett. **107**, 103902 (2011).
- [16] C. I. Hancox, S. C. Doret, M. T. Hummon, R. V. Krems, and J. M. Doyle, Phys. Rev. Lett. **94**, 013201 (2005).
- [17] R. V. Krems, J. Kłos, M. F. Rode, M. M. Szczeniński, G. Chałasiński, and A. Dalgarno, Phys. Rev. Lett. **94**, 013202 (2005).
- [18] M.-J. Lu, V. Singh, and J. D. Weinstein, Phys. Rev. A **79**, 050702 (2009).
- [19] M. Lu, N. Q. Burdick, S. H. Youn, and B. L. Lev, Phys. Rev. Lett. **107**, 190401 (2011).
- [20] M. Lu, N. Q. Burdick, and B. L. Lev, Phys. Rev. Lett. **108**, 215301 (2012).
- [21] S. Kotochigova and A. Petrov, Phys. Chem. Chem. Phys. **13**, 19165 (2011).
- [22] A. Petrov, E. Tiesinga, and S. Kotochigova, arXiv:1203.4172 (2012).
- [23] K. Aikawa, A. Frisch, M. Mark, S. Baier, A. Rietzler, R. Grimm, and F. Ferlaino, Phys. Rev. Lett. **108**, 210401 (2012).
- [24] L. P. Parazzoli, N. J. Fitch, P. S. Żuchowski, J. M. Hutson, and H. J. Lewandowski, Phys. Rev. Lett. **106**, 193201 (2011).
- [25] V. Kokorouline, R. Santra, and C. H. Greene, Phys. Rev. Lett. **90**, 253201 (2003).
- [26] R. Santra and C. H. Greene, Phys. Rev. A **67**, 062713 (2003).
- [27] T. Kuwamoto, K. Honda, Y. Takahashi, and T. Yabuzaki, Phys. Rev. A **60**, R745 (1999).
- [28] S. Uetake, A. Yamaguchi, S. Kato, and Y. Takahashi, Appl. Phys. B **92**, 33 (2008).
- [29] A. Yamaguchi, S. Uetake, and Y. Takahashi, Appl. Phys.

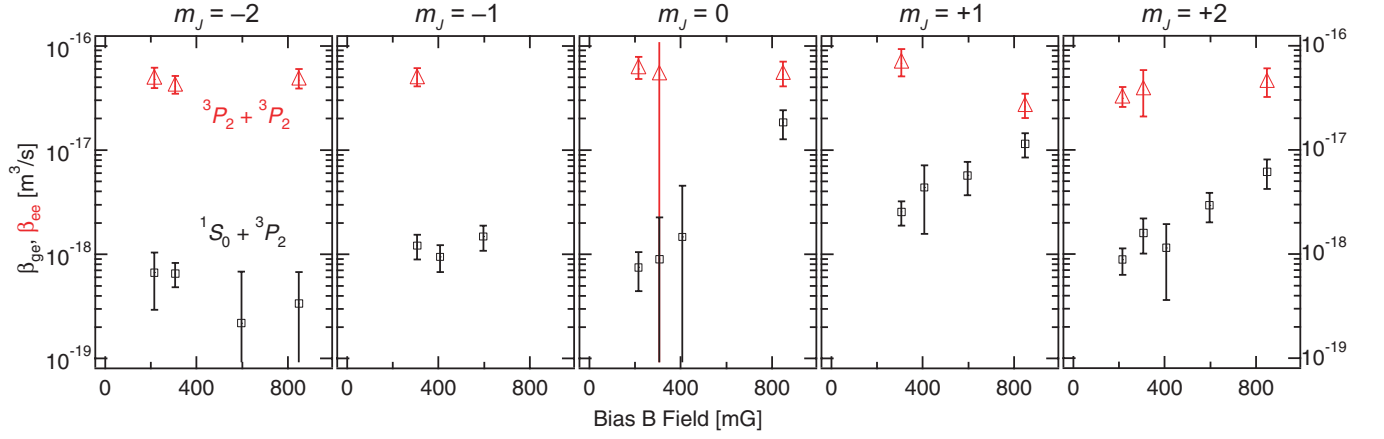


FIG. 3. (color online) Inelastic decay rate for all m_J states in various external magnetic fields. Red triangles show inelastic decay rate β_{ee}^{in} of 3P_2 - 3P_2 collision system. Essentially no spin-dependence is observed. Black squares show those of 1S_0 - 3P_2 collision system (β_{ge}^{in}). Significant spin- and magnetic-field-dependence is observed.

- B **91**, 57 (2008).
- [30] A. Yamaguchi, S. Uetake, S. Kato, H. Ito, and Y. Takahashi, New Journal of Physics **12**, 103001 (2010).
- [31] T. Ido and H. Katori, Phys. Rev. Lett. **91**, 053001 (2003).
- [32] O. J. Luiten, M. W. Reynolds, and J. T. M. Walraven, Phys. Rev. A **53**, 381 (1996).
- [33] R. deCarvalho and J. Doyle, Phys. Rev. A **70**, 053409 (2004).
- [34] M. Yan, R. Chakraborty, P. G. Mickelson, Y. N. M. de Escobar, and T. C. Killian, arXiv:0905.2223 (2009).
- [35] T. V. Tscherbul, (private communication).
- [36] S. Sugawa, S. Kato, S. Uetake, and Y. Takahashi, (in preparation).